Unlimited Release
UC-71

An Evaluation of Three Partially Volatile Neutron Shields for High-Performance Shipping Casks

H. J. Rack, H. S. Pearson
Transportation Technology Center

Prepared by Sandia Laboratories, Albuquerque, New Mexico 87185 and Livermore, California 94550 for the United States Department of Energy under Contract DE-AC04-76DP00789.

PRINTED FEBRUARY 1981

Issued by Sandia Laboratories, operated for the United States Department of Energy by Sandia Corporation.

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161

NTIS price codes
~~Printed copy: $6.00~~
Microfiche copy: A01

TTC-0149
SAND80-2303
Unlimited Release
Printed February 1981

AN EVALUATION OF THREE PARTIALLY VOLATILE NEUTRON SHIELDS FOR HIGH-PERFORMANCE SHIPPING CASKS*

H. J. Rack
Mechanical Metallurgy Division
Sandia National Laboratories†
Albuquerque, New Mexico 87185

H. S. Pearson
Applied Technical Services‡
Marietta, Georgia

ABSTRACT

The thermal stability and mechanical behavior of three partially volatile candidate neutron shield materials have been evaluated. The results indicate that silicone based rubbers, impregnated with elemental boron or boron carbide, Boro-silicone 236 and Bisco NS-I respectively are more thermally stable than are borated beechwoods, e.g., Permali JN. Mechanical property measurements indicated however that the compressive strength of the borated beechwood is 10 to 48 times higher than that of the silicone-based rubbers. The compressive strengths of the borated beechwood and boron carbide impregnated silicone rubber were substantially more sensitive to test temperature than was the compressive strength of the boron impregnated silicone rubber. Finally the compressive strengths and energy absorbing capability of the boron impregnated silicone rubber is not affected by prior thermal exposure at 425K for 1000h.

*This work was supported by the U. S. Department of Energy (DOE), under Contract DE-AC04-76-DP00789.

†A U. S. Department of Energy Facility.

‡Present address: Lockheed-Georgia Company, Marietta, Ga.

ACKNOWLEDGMENT

The authors wish to acknowledge the assistance of T. Massis, J. Rowe and S. L. Erickson in performing the thermo-gravometric analysis, weight loss and chemical analysis experiments reported herein.

CONTENTS

ILLUSTRATIONS

TABLES

INTRODUCTION

Many spent-fuel shipping casks in use throughout the world utilize water or ethylene glycol/water mixtures as the neutron shielding material. In either case the shield material is usually contained in a cavity near the outer surface of the cask. As such, the probability of retaining the liquid neutron shield in a severe accident is low. For such casks, the designer usually is required by his certificating authority to assume in his safety analysis that the shield material is lost during an accident. The radiation level external to the package following an accident must not exceed that which is allowed by regulation. In the IAEA Safety Series No. 6, paragraphs 229 and 532, an increase in external dose from 10 mrem/h, 2 m from the external surface of the package to 1000 mrem/h, 1 m from the surface of the package is allowed following the "tests for demonstrating ability to withstand accident conditions in transport."

At least three problems associated with the use of volatile liquids as neutron shielding materials in shipping casks have been identified. First, for casks such as might be needed for the shipment of high-burnup, short-cooled spent fuel from liquid metal fast breeder reactors (LMFBR), the external dose rate following exposure to the accident-related tests may exceed that allowed by the regulations if all of the neutron shielding material is lost. Specifically, this occurs if the design of the cask has been optimized about a balanced shielding of both gamma rays and neutrons. Complete loss of the neutron shield following an accident requires that the designer add extra gamma shielding. This results in significant increases in cask weight and overall dimensions. Second, the use of volatile neutron shields may often complicate the packaging design and operation, since expansion volumes and pressure relief systems must be provided to prevent over-pressurization during normal use and exposure to fire environments. Third, irrespective of the material being carried, the loss of all the neutron shielding material in an accident results in a more difficult and potentially more hazardous accident recovery procedure.

Recently a study was initiated to evaluate the feasibility of fabricating neutron shields for spent fuel shipping casks using commercially available materials which would retain at least part of their neutron shielding capability following exposure to a 1075K, 30-minute thermal test. This concept utilizes interstitially located layers of a neutron shielding material and metallic heat transfer fins. Various materials which could either be preformed to specific shapes and inserted into the neutron shield cavities or could be "injected" into the cavities and solidified in-situ were examined. These preliminary evaluations resulted in the selection of three candidates: (a) a boron loaded silicone rubber, Boro-silicone rubber 236, a product of Reactor Experiments, Inc.: (b) a boron carbide loaded silicone rubber Bisco NS-I, a product of Brand Industrial Services Inc., and (c) a boron oxide impregnated cross-laminated beechwood, Permali JN, a product of the Permali Industrial Group. This report describes studies performed to assess the thermal stability and mechanical behavior of these three candidate materials.

EXPERIMENTAL PROCEDURES

Materials

The three candidate neutron shield materials examined were received in the form of 5.1 cm thick x 51 cm square blocks. Table 1 lists the H, C and B contents of the as-received material. The Boro-silicone 236 contained 1.6 wt. pct. B in the form of elemental boron, the Bisco NS-I contained 16.1 wt. pct. B in the form of B_4C, and the Permali JN contained 2.1 wt. pct. in the form of B_2O_3. In addition the Boro-silicone 236 contained approximately 16 wt. pct. Al apparently as an aluminum oxide filler.

Thermal Stability

The thermal stability of the Boro-silicone 236, Bisco NS-I and Permali JN was initially evaluated using thermo-gravometric analysis (TGA), with more detailed long time stability being established using standard isothermal weight change (loss) procedures. The former (TGA) procedure involved a measurement of the percent weight loss which occurred during isochronel (constant rate) heating of the test sample. Atmosphere control during heating also made it possible to consider the possible influence of an environment, e.g., air versus argon, on the material's thermal stability.

Results of the TGA measurements were used to establish the temperatures for the isothermal weight loss measurements. Duplicate weight change samples 1.27 cm sq. x 5.1 cm high, having a surface to volume ration of 0.177, were then exposed at selected temperatures in an air environment for times up to 2024 h.

Mechanical Properties

Triplicate tensile samples, as shown in Fig. 1, were used to evaluate the tensile properties of Permali JN. These samples contained 10-11 plies

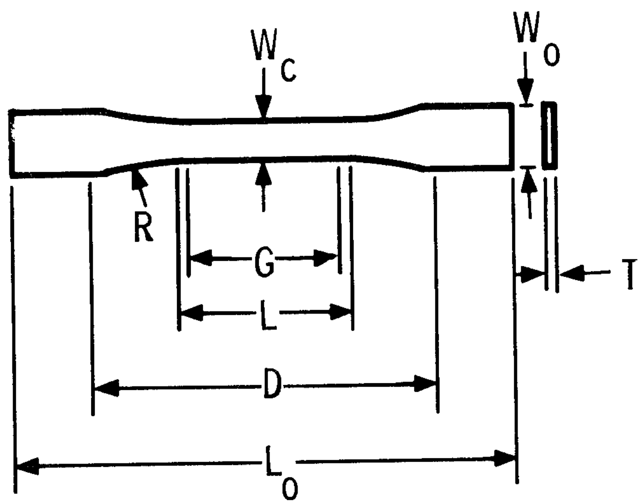
Table 1

Chemical Analysis of Partially Volatile Candidate Neutron Shield Materials Prior to Thermal Exposure

Candidate Material	Weight Percent		
	H	C	B[a]
Boro-silicone 236[b]	4.5	11.4	1.6
Bisco NS-I	3.3	18.1	16.1
Permali JN	5.6	43.0	2.1

(a) Boron present as elemental boron, B_4C and B_2O_3 for Boro-silicone 236, Bisco NS-I and Permali JN, respectively.

(b) Also contained approximately 16 wt. pct. Al as aluminum oxide.



DIMENSIONS (mm):

W_C	19	D	114.3
W_O	28.7	L_O	246.4
G	50	R	76.2
L	57.2	T	12.7

Fig. 1. Tensile Configuration for Permali JN

and were removed from both the LT and TL ply orientations, the latter as defined in Fig. 2. Measurements of the compressive properties of Permali JN, as well as Boro-silicone 236 and Bisco NS-I utilized samples having a length to diameter/side ratio of 2 with the length being 2.54 cm. The compressive properties of the three candidate neutron shield materials were determined at a strain rate of 10^{-4} sec^{-1} over the temperature range 233 to 473K, with all specimens having been soaked at temperature for at least 0.5h prior to testing.

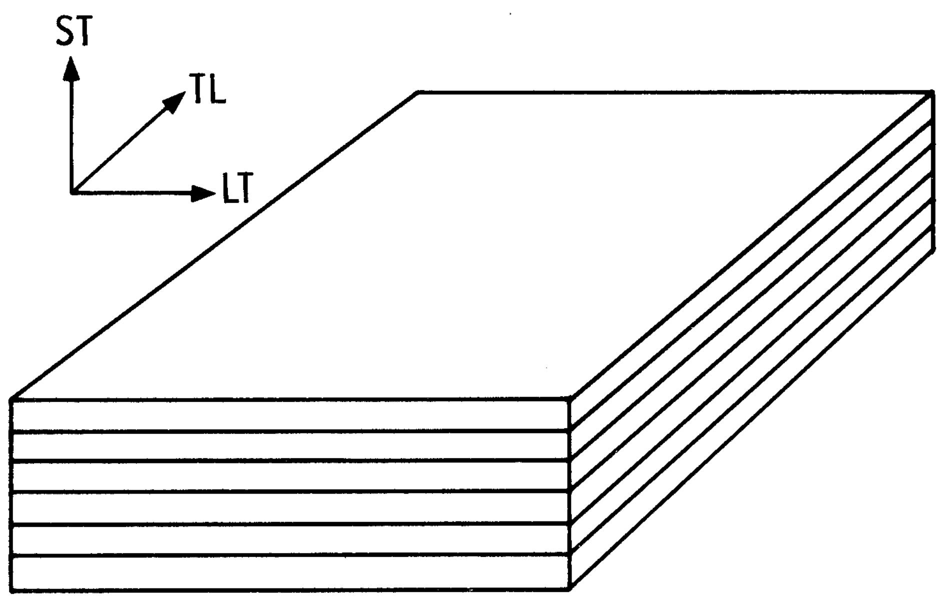


Fig. 2. Sample Orientation for Permali JN

This Page Intentionally Left Blank

EXPERIMENTAL RESULTS AND DISCUSSION

Thermal Stability

Fig. 3 presents the results of the thermo-gravometric analysis of the three candidate materials. These results indicate that the silicone based materials, Boro-silicone 236 and Bisco NS-I, are stable to $\sim$ 475K, with a gradual weight loss occurring above this temperature. In contrast Permali JN is unstable at all temperatures above ambient, with significant instability beginning at approximately 475K. This latter degradation is so severe that at temperatures exceeding 975K essentially all of the material has been volatilized.

Another measure of the relative thermal stability of the candidate materials can be obtained using the thermo-gravometric analysis results by establishing the temperature to which the materials must be heated at a constant heating rate to achieve a pre-selected weight loss. Table 2 presents such a comparison for a 5 percent weight loss. This table shows that Bisco NS-I is the most stable material under both inert (argon) and oxidizing (air) environments. It should be noted, however, that the thermal stability of Bisco NS-I appears to be more sensitive to cover-gas environment than is Boro-silicone 236. The temperature for attaining a 5 percent weight loss in Boro-silicone 236 is essentially independent of the cover-gas, while that for Bisco NS-I decreases by $\sim$ 50K by changing from an inert (argon) atmosphere to an oxidizing (air) environment.

The final method used to assess the long term thermal stability of the candidate neutron shield materials involved isothermal weight-loss measurements in an oxidizing (air) environment. These results, Fig. 4, show that, contrary to the thermo-gravometric analysis observations, long term exposure of Boro-silicone

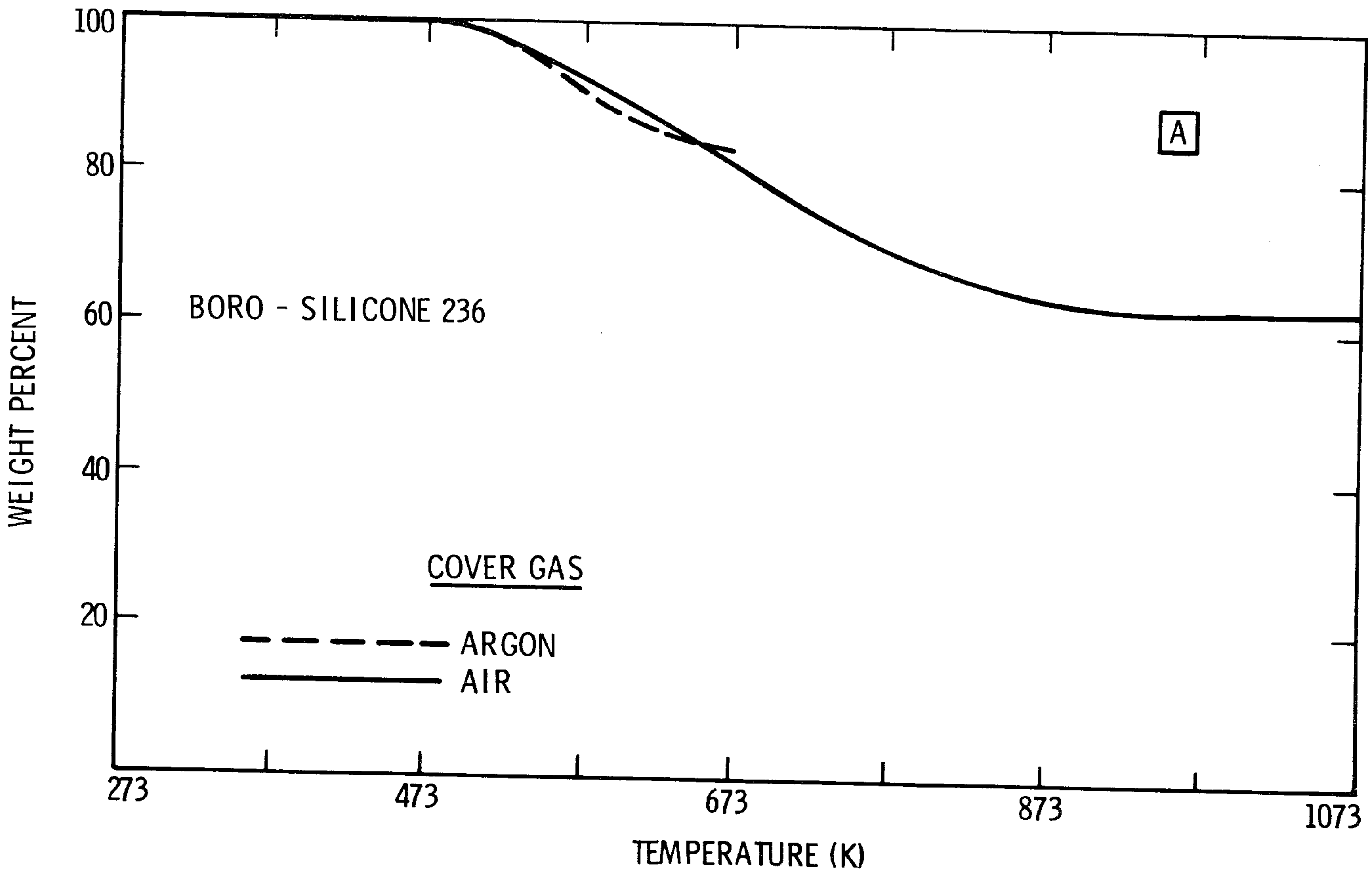
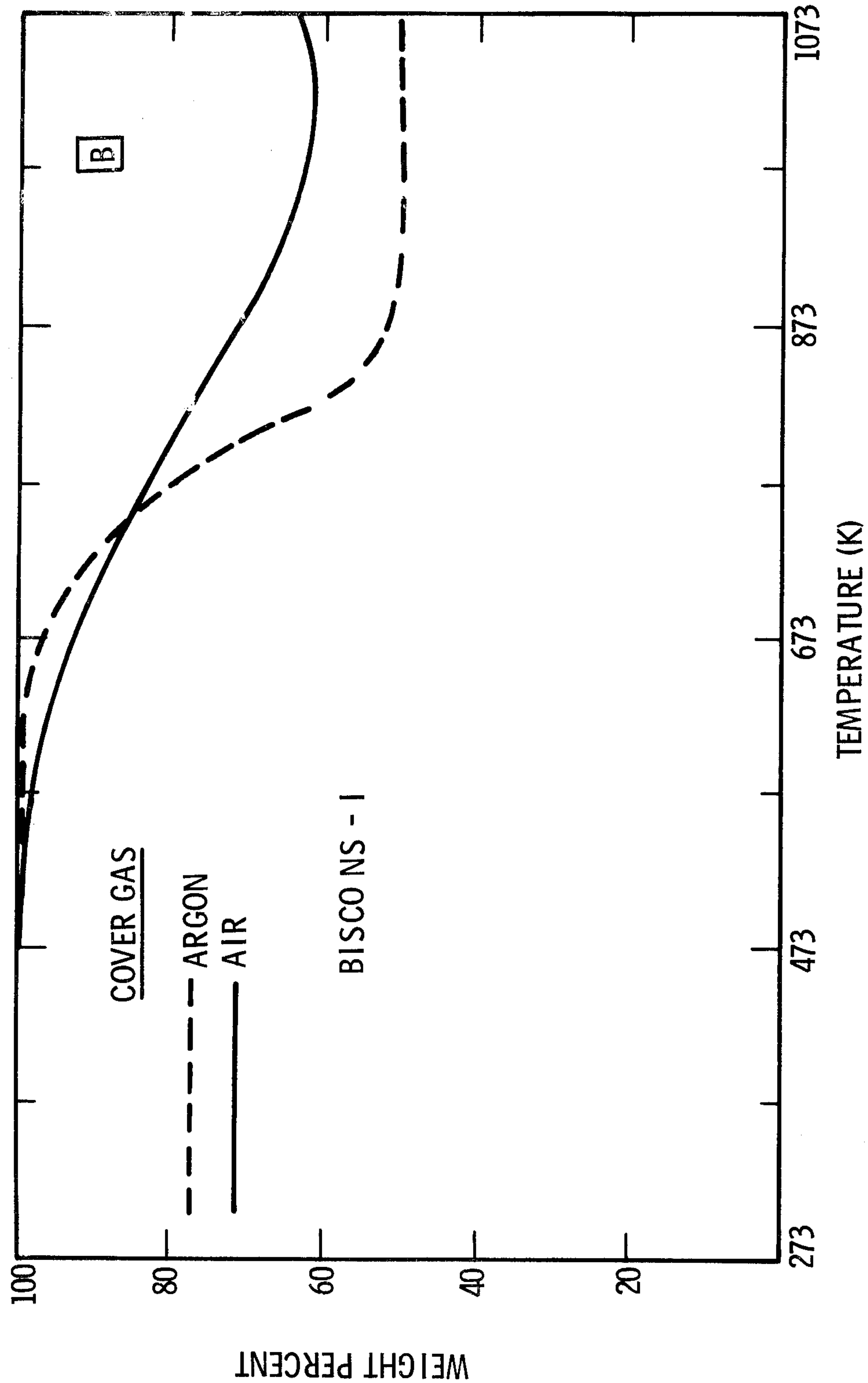
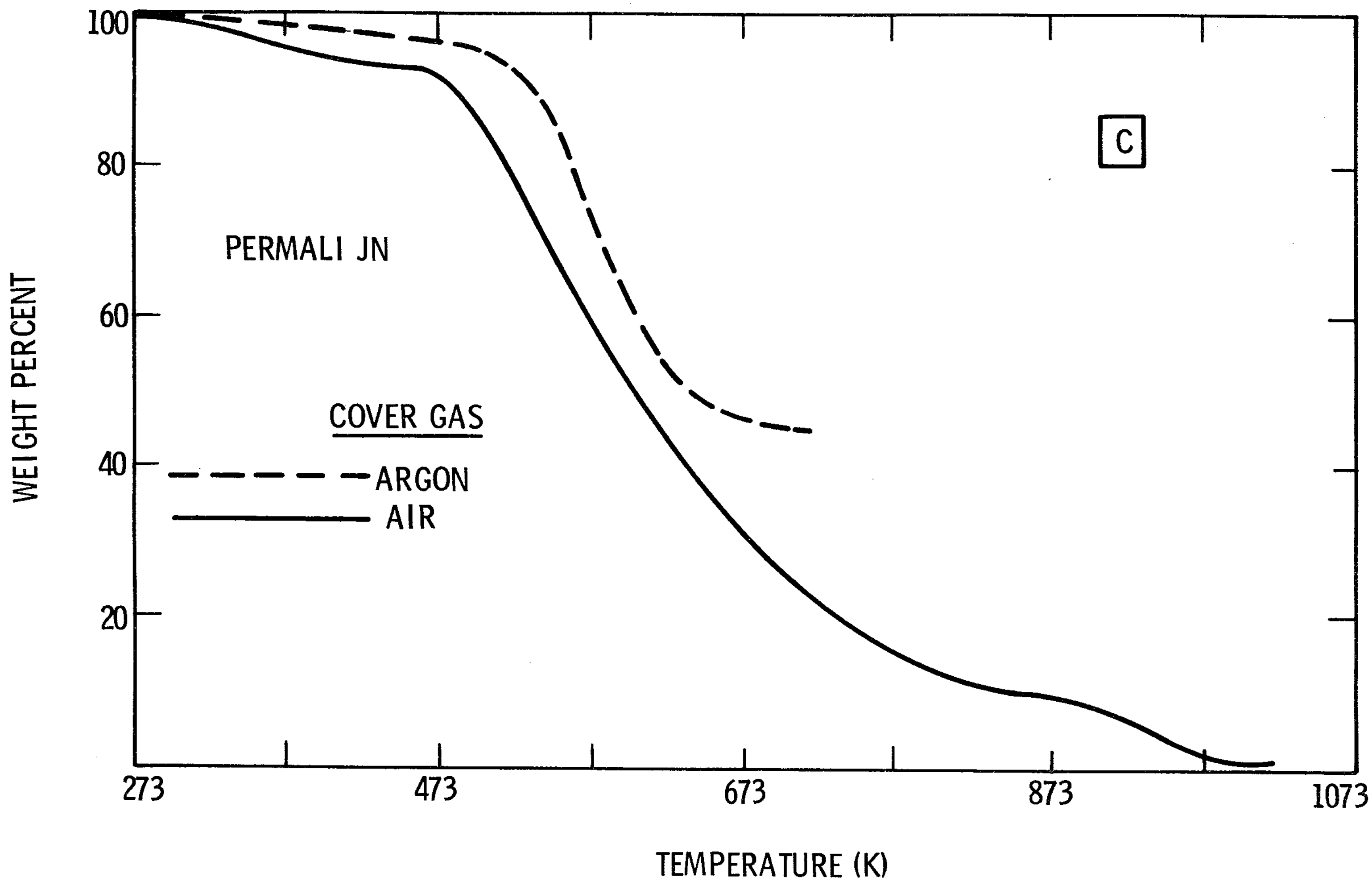


Fig. 3. Thermo-gravometric analysis of candidate neutron shield materials. Heating rate: 10K/min.



WEIGHT PERCENT
100
80
60
40
20
273
473
673
873
1073
TEMPERATURE (K)
COVER GAS
ARGON
AIR
BISCO NS - I
B



100
80
60
40
20
WEIGHT PERCENT
273
473
673
873
1073
TEMPERATURE (K)
PERMALI JN
COVER GAS
ARGON
AIR
C

Table 2

Temperature for Achieving 5 Percent Weight Loss in Constant Heating Rate (10K/min) Evaluation of Candidate Neutron Shield Materials

Candidate Material	Air Atmosphere Temperature (K)	Argon Atmosphere Temperature (K)
Boro-silicone 236	552	547
Bisco NS-I	641	694
Permali JN	388	503

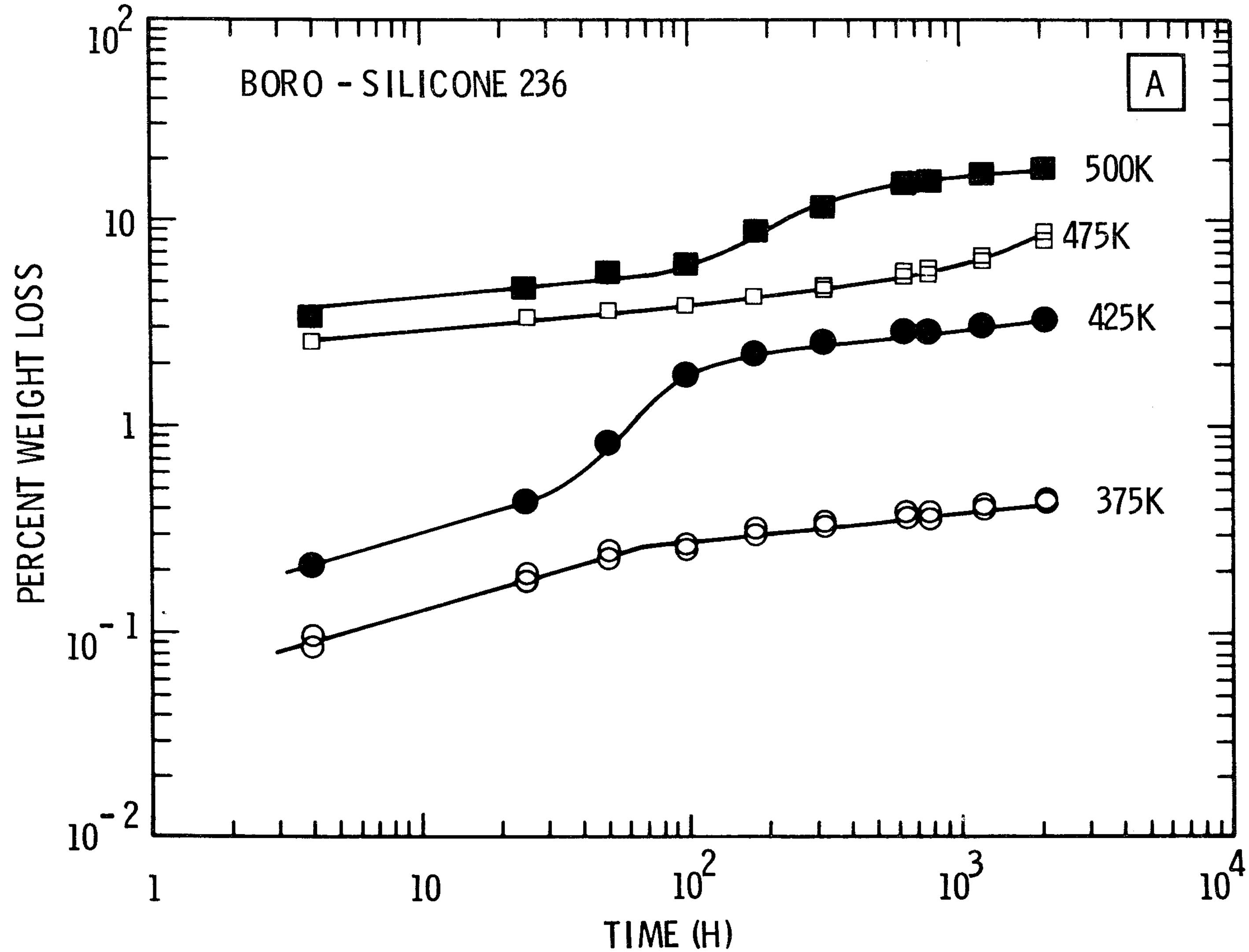
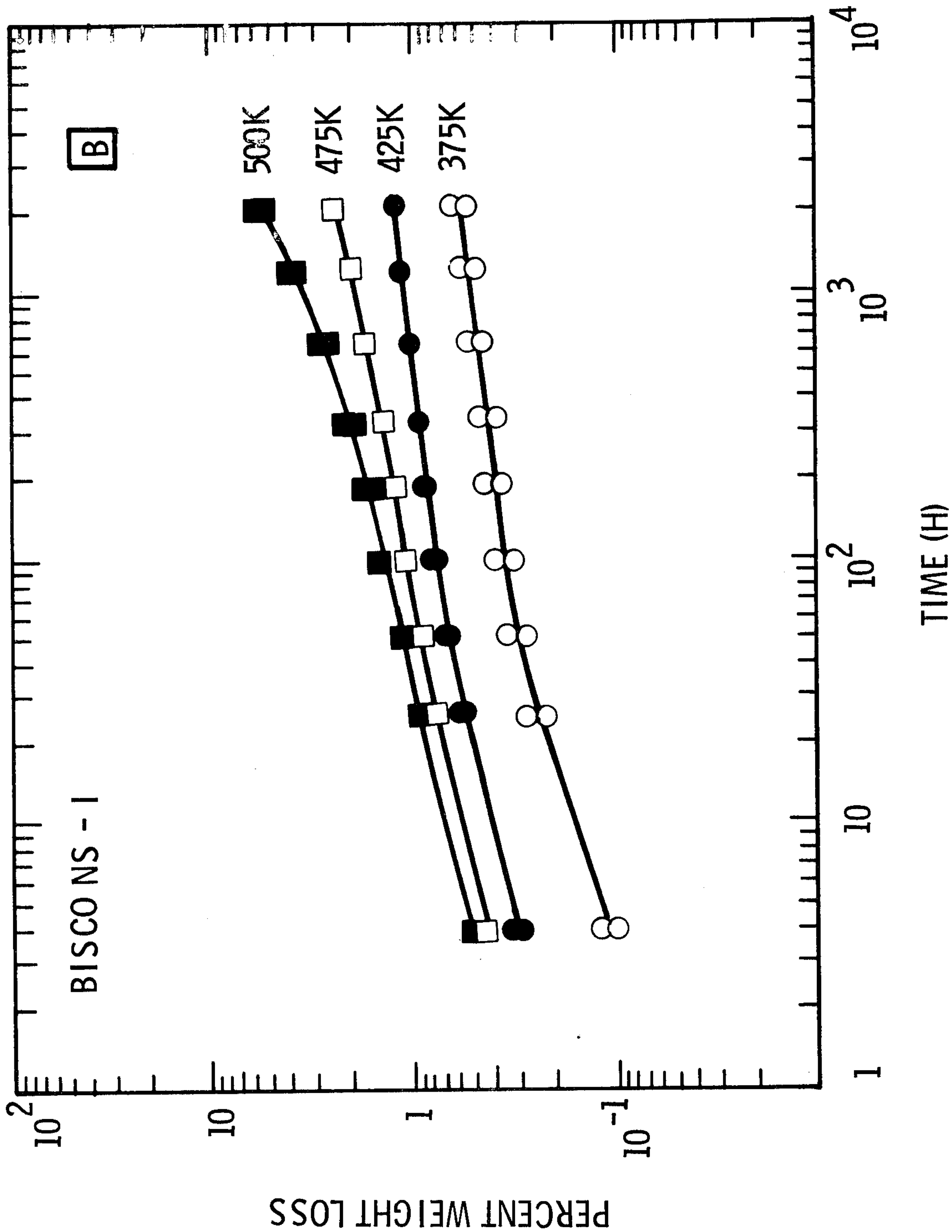
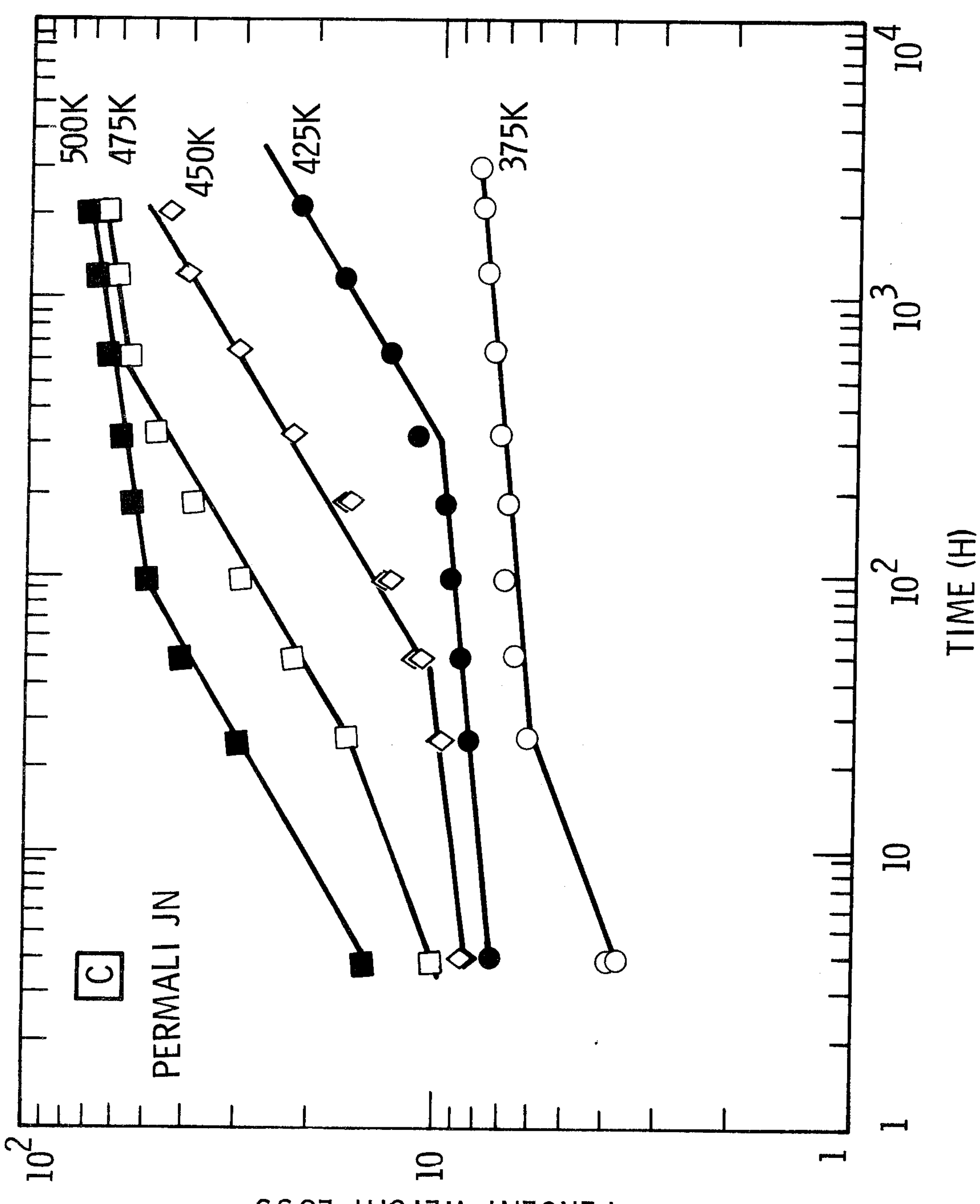


Fig. 4. Isothermal weight loss for (a) Boro-silicone 236, (b) NS-I and (c) Permali JN. Temperatures as indicated.


BISCONS - I
B
500K
475K
425K
375K
PERCENT WEIGHT LOSS
10^{2}
10
1
10^{-1}
TIME (H)
1
10
10^{2}
10^{3}
10^{4}


C
PERMALI JN
500K
475K
450K
425K
375K
TIME (H)
PERCENT WEIGHT LOSS
1
10
10^2
10^3
10^4

236 and Bisco NS-I below 475K can lead to degradation, although the percentage weight losses observed were quite low, typically less than 2 percent after 2024 h exposures. Fig. 5 presents a direct comparison, at constant temperature, of the three candidate neutron shield materials. In each case the silicone based materials were again more stable than the Permali JN. These results also indicate that inclusion of time dependent aging effects leads to a decrease in the maximum continuous operating temperature for the three candidate materials relative to that which would have been obtained by only considering the thermal-gravometric analysis results. Table 3 presents such a comparison for the three materials at the 5 percent weight loss level. The isothermal observations as shown in Table 3 also tend to reduce the differences between the two silicone based materials. Finally, chemical analysis, Table 4, indicated that the weight losses observed after isothermal exposure in an air environment appeared to be related to a reduction in the hydrogen content of the three candidate neutron shield materials.

Mechanical Behavior

The results of the room temperature tensile and compressive measurements of Permali JN are summarized in Table 5. Tensile loading of this material results in an elastic failure, the final failure mode involving tearing of the beechwood cross-plies as shown in Fig. 6. This failure mode is in contrast with the shear type failures observed when Permali JN was compressively loaded in the LT direction, Fig. 7. Table 5 shows that the tensile and compressive properties of the Permali JN examined in this study were, contrary to manufacturing specification for a cross-ply laminate, functions of orientation. Indeed the observed orientations effects were quite large and would have to be considered in any neutron shield design.

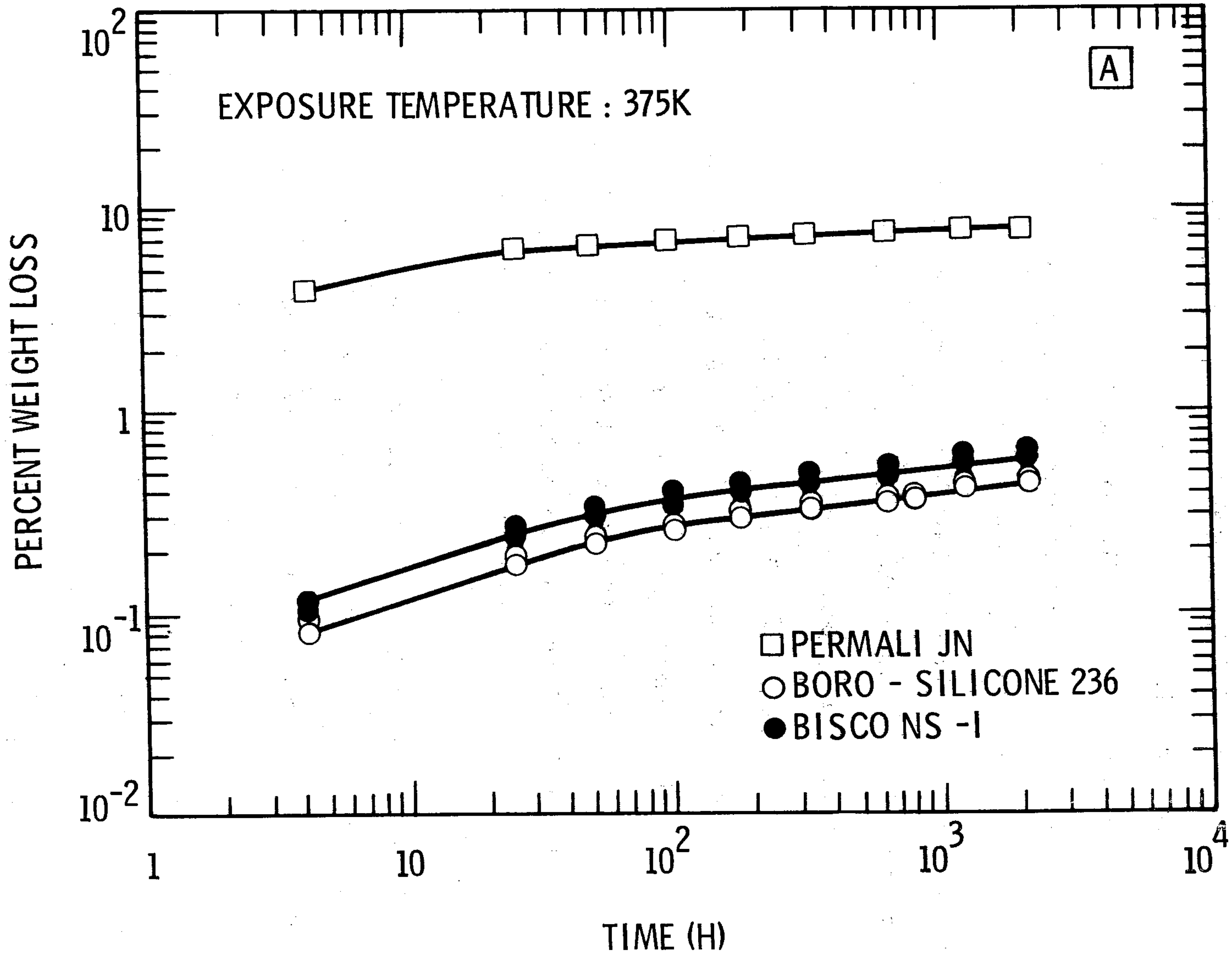
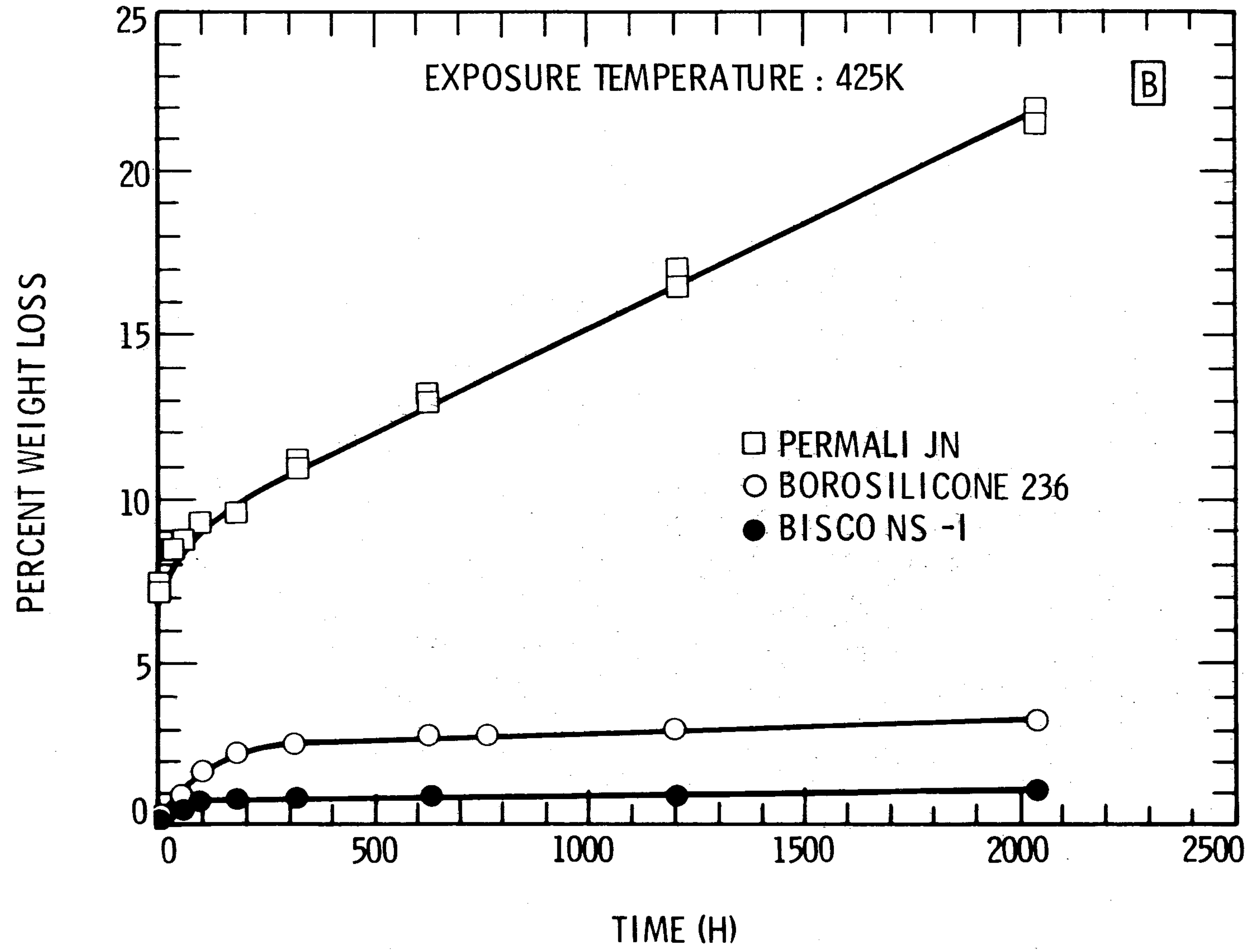


Fig. 5. Comparison of isothermal weight loss measurements for Boro-silicone 236, NS-I and Permali JN at (a) 375K, (b) 425K and (c) 475K.


EXPOSURE TEMPERATURE : 425K
B
PERMALI JN
BOROSILICONE 236
BISCO NS -1
PERCENT WEIGHT LOSS
TIME (H)
0
5
10
15
20
25
0
500
1000
1500
2000
2500

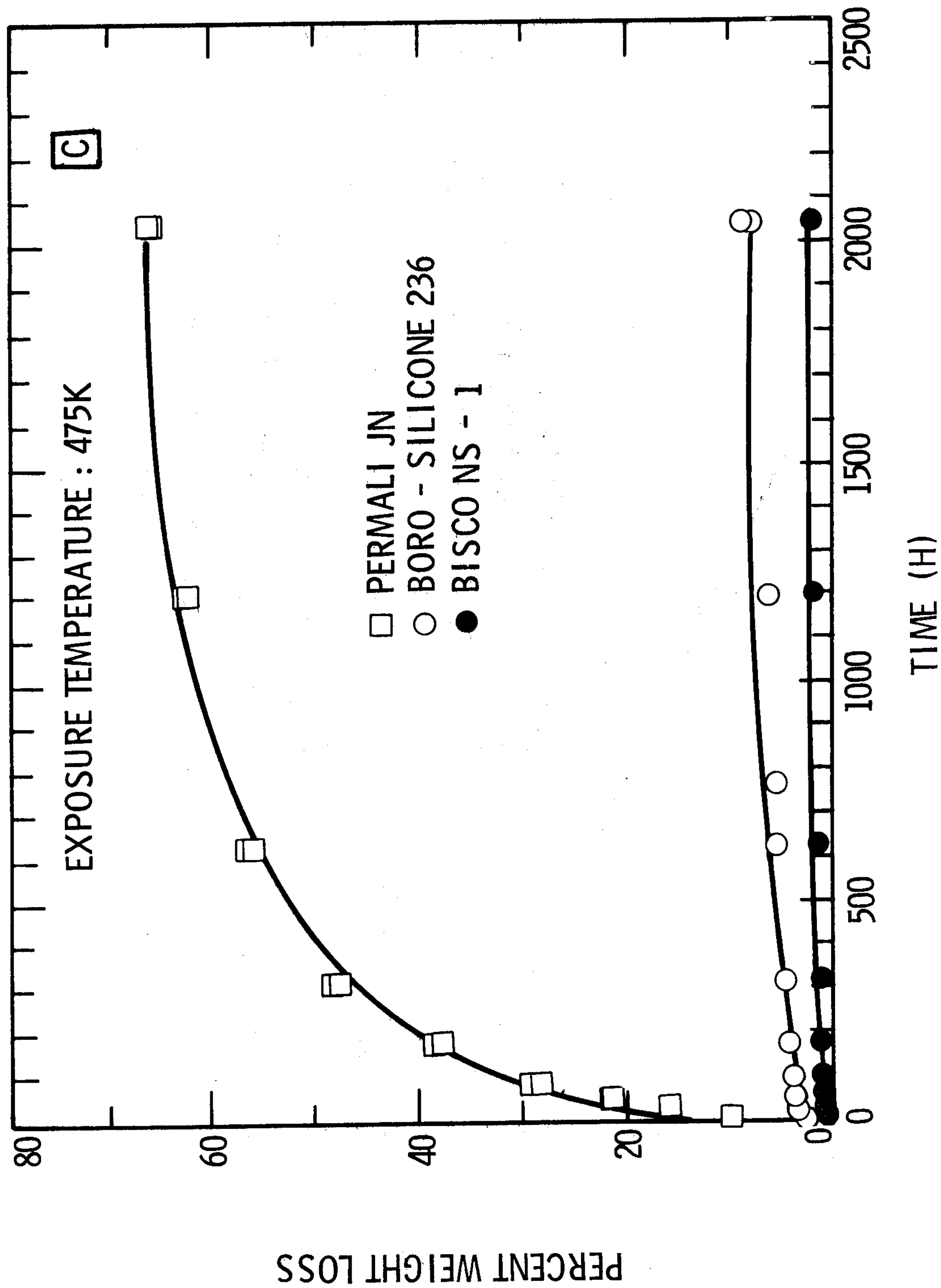

EXPOSURE TEMPERATURE : 475K
C
PERMALI JN
BORO - SILICONE 236
BISCO NS - 1
PERCENT WEIGHT LOSS
0
20
40
60
80
TIME (H)
0
500
1000
1500
2000
2500

Table 3

Temperature for Achieving 5 Percent Weight Loss From TGA and Isothermal Measurements+

Candidate Material	TGA Temperature (K)	Isothermal Temperature (K)
Boro-silicone 236	552	~ 450
Bisco NS-I	641	~ 475
Permali JN	388	< 375

+Air atmosphere

Table 4

Chemical Analysis of Partially Volatile Candidate Neutron Shield Materials Following Thermal Exposure+

Candidate Material	Exposure Temperature (K)	Weight Percent		
		H	C	B
Boro-silicone 236	298	4.5	11.4	1.6
	375	4.5	11.2	1.9
	425	4.1	10.9	1.6
	475	4.1	11.5	1.3
	500	3.2	10.6	1.7
Bisco NS-I	298	3.3	18.1	16.1
	375	3.2	17.8	17.2
	425	3.1	18.2	16.8
	475	3.0	17.6	17.3
	500	3.0	17.7	17.7
Permali JN	298	5.6	43.0	2.1
	375	5.5	44.2	2.1
	425	4.7	45.0	2.5
	450	3.6	44.6	3.5
	475	2.3	43.8	5.3
	500	1.8	42.4	6.2

+Exposed 2024 h at indicated temperatures

Table 5

Room Temperature Mechanical Properties of Permali JN[a]

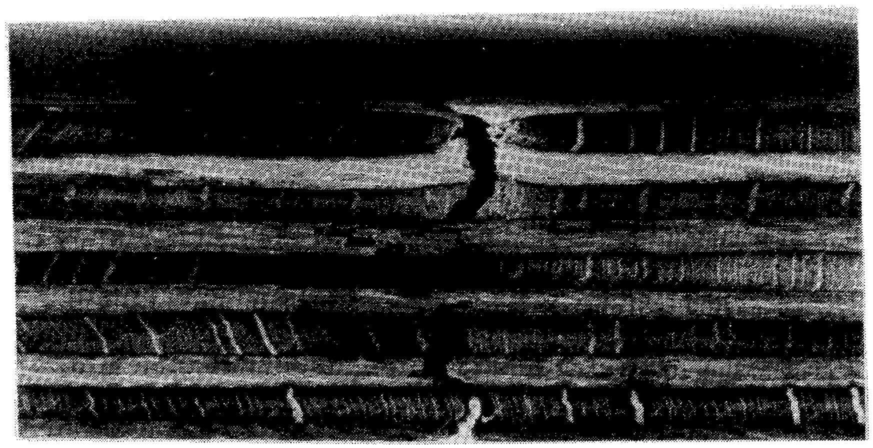
	Orientation	$\sigma_{0.5}$[b] (MPa)	σ_u[c] (MPa)	Y[d] (GPa)
Tension	LT	--	86	16.1
	TL	--	49	12.1
Compression	LT	91	124	11.0
	ST	112	190	2.7

(a) Strain rate: 10^{-4} sec^{-1}, as-received condition.

(b) $\sigma_{0.5}$: flow stress at 0.005 strain

(c) σ_u: ultimate strength

(d) Y: Young's modulus



Tensile Axis

Fig. 6. Tensile Failure in Permali JN

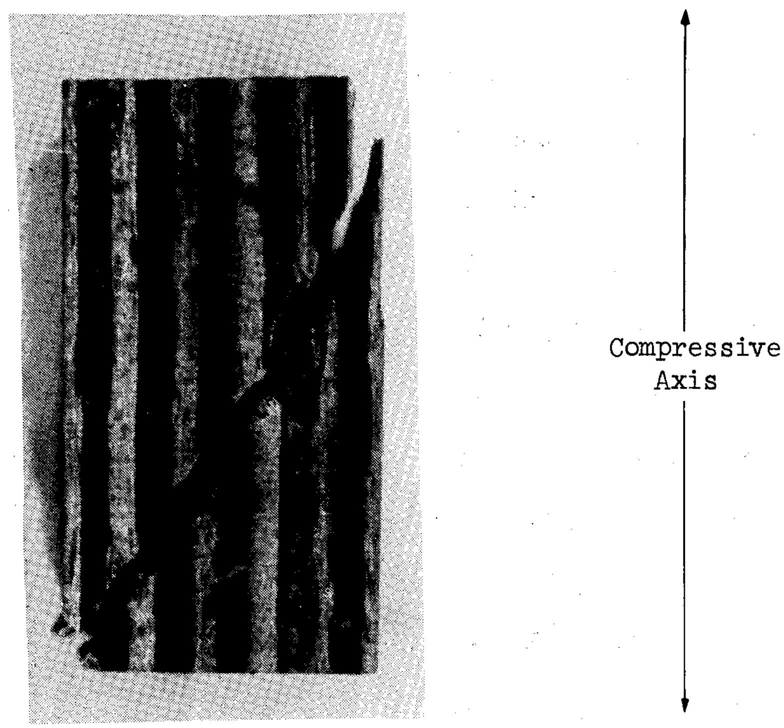


Fig. 7. Compressive Failure in Permali JN.
LT Orientation.

Further examination showed that the compressive properties of Permali JN were also quite sensitive to test temperature, Fig. 8. For example, decreasing the test temperature from 400K to 233K tripled the compressive strength of Permali JN. Finally, there appeared to be some increase in the compressive strength of Permali JN when the test temperature was increased from 400 to 450K.

Comparison of the results shown in Fig. 9 for Boro-silicone 236 and Bisco NS-I with those for Permali JN, Fig. 8, indicated that the compressive strengths of the silicone based rubbers were substantially below those of Permali JN. For example the room temperature compressive strength of Bisco NS-I was approximately 12 MPa and that of Boro-silicon 236, 2.5 MPa, factors of 10 and 48 below that of Permali JN. The compressive strength of Boro-silicone 236, as compared to both Permali JN and Bisco NS-I, was relatively insensitive to test temperature. Finally, Fig. 9 shows that prior thermal aging, in this case for 1000h at 425K, had little effect on the compressive behavior of Boro-silicone 236 and only effected that of Bisco NS-I at 233K.

One final comparison between the two silicone-based condidate neutron shield materials is presented in Fig. 10. Here the energy absorbed during compressive straining to a total strain of 15 percent has been plotted against test temperature. These results, which are typical of all strain offsets examined, indicate that (a) the energy absorption capability of these silicone rubbers was essentially independent of test temperature, (b) Boro-silicone 236 will absorb approximately twice the energy to a fixed strain than will Bisco NS-I, and (c) that aging, again for 1000h at 425K, does not effect the energy absorption capability of either Boro-silicone 236 or Bisco NS-I.

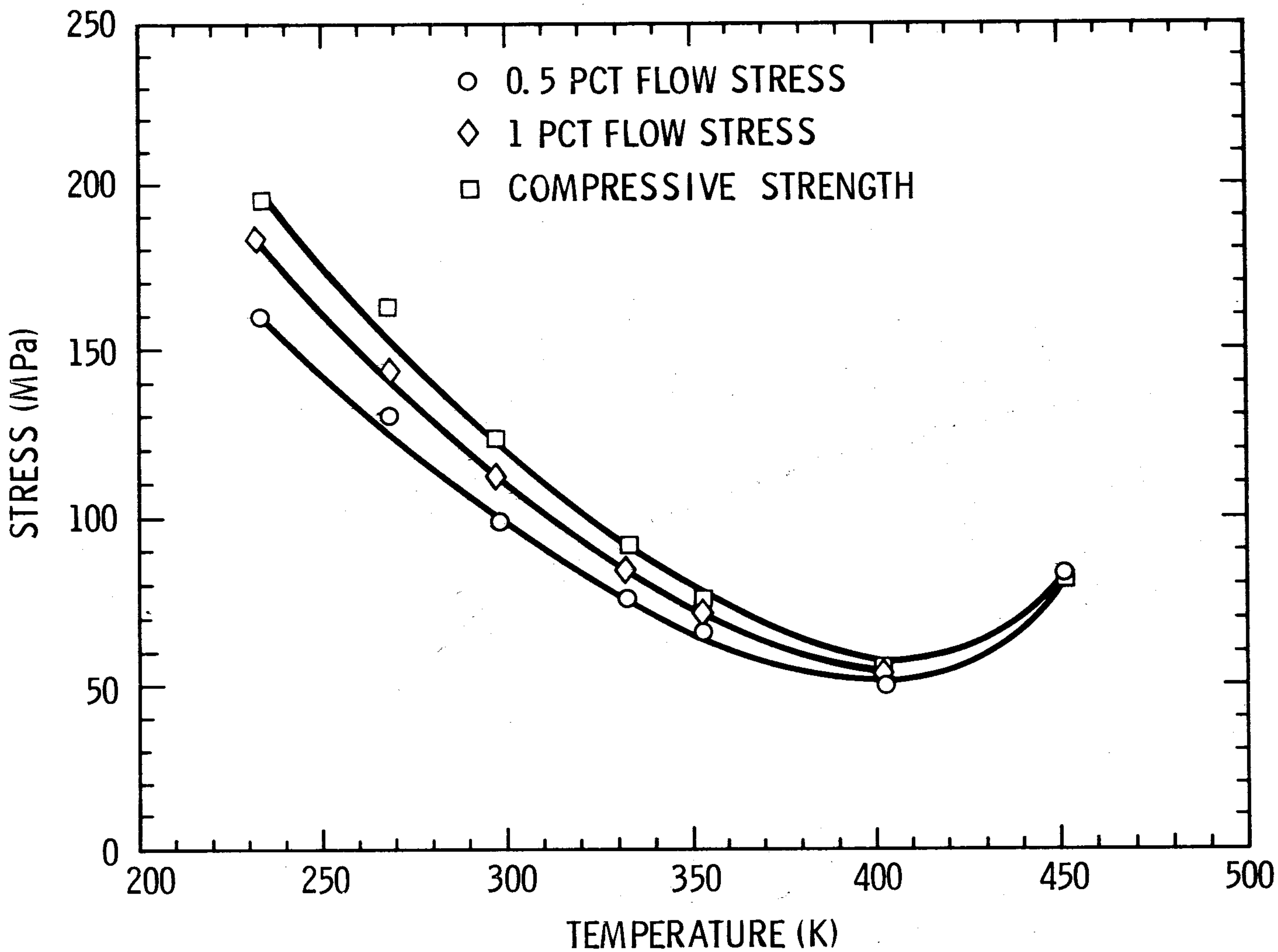


Fig. 8. Influence of Test Temperature on the Compressive Properties of Permali JN - LT Orientation

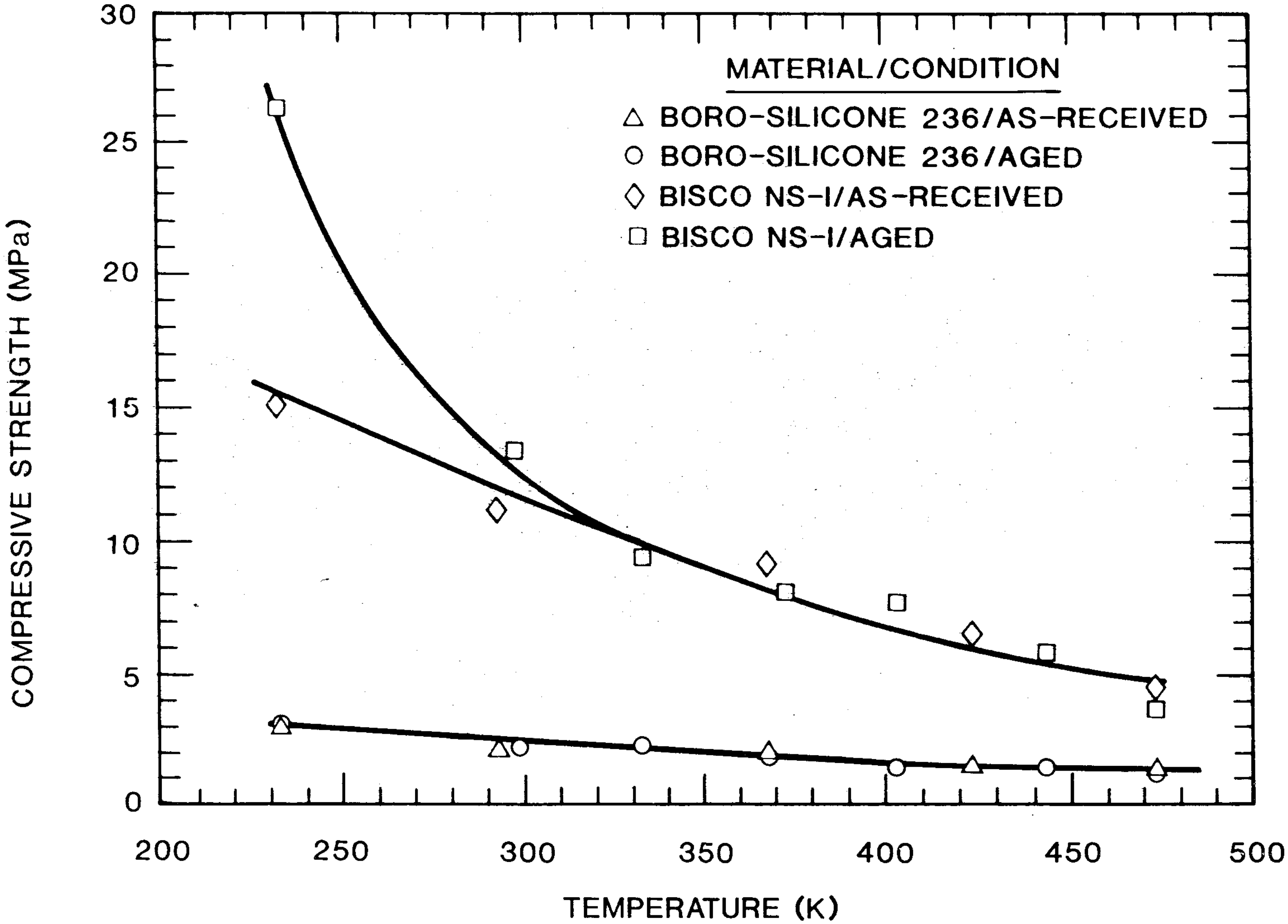


Fig. 9. Influence of Test Temperature on the Compressive Strength of Boro-Silicone 236 and Bisco NS-I.

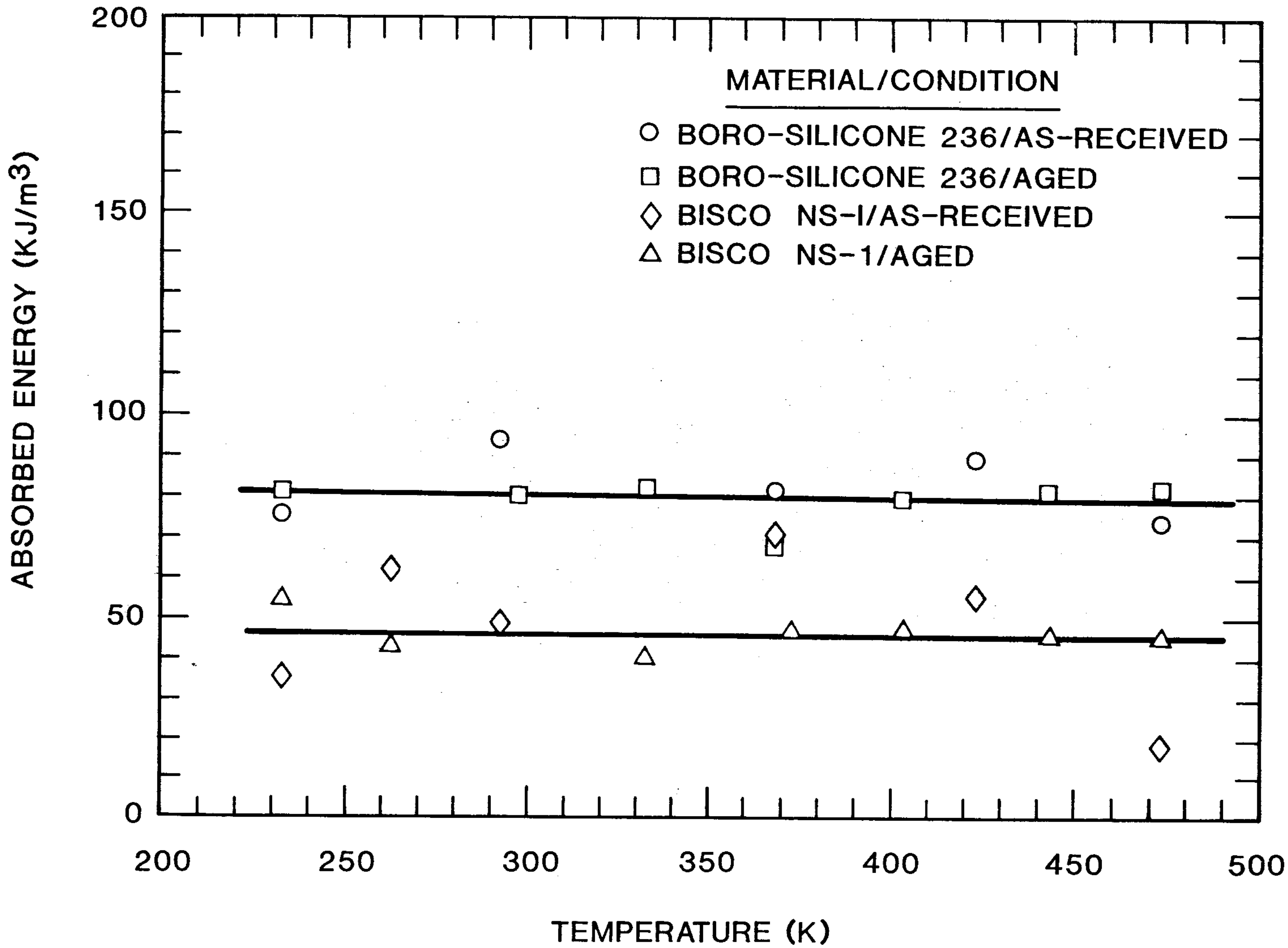


Fig. 10. Influence of Test Temperature on Absorbed Energy of Boro-Silicone 236 and Bisco NS-I.

This Page Intentionally Left Blank

SUMMARY AND CONCLUSIONS

The present investigation of three partially volatile candidate neutron shield materials has shown that silicone-based rubbers, e.g., Boro-silicone 236 and Bisco NS-I, are more thermally stable than borated beechwood, Permali JN. Under oxidizing (air) conditions these materials have upper continuous operating temperatures of 450K (Boro-silicone 236), 475K (Bisco NS-I) and < 375K (Permali JN).

Mechanical property measurements have shown that the compressive strength of Permali JN is typically 10-48 times higher than either Boro-silicone 236 or Bisco NS-I. In addition, the compressive strengths of Permali JN and Bisco NS-I increase markedly with decreasing temperature, while that of Boro-silicone 236 is relatively insensitive to temperature. Finally it has been shown that Boro-silicone 236 will absorb approximately twice as much energy during compressive straining than will Bisco NS-I and that prior thermal exposure (aging) and test temperature has little effect on this energy absorbing capability.

This Page Intentionally Left Blank

DISTRIBUTION:
Unlimited Release

U. S. Department of Energy
Technical Information Center
Attn: DOE/TIC-4500-R67 UC-71 (166)

Allied-General Nuclear Services
P. O. Box 847
Barnwell, SC 29812
Attn: P. Highberger

Atomics International
P. O. Box 309
Canoga Park, CA 91304
Attn: K. Foster

Battelle Columbus Laboratories
Office of Nuclear Waste Isolation
505 King Ave.
Columbus, OH 43201
Attn: R. Peterson

Battelle Memorial Institute
Pacific Northwest Laboratory
P. O. Box 999
Richland, WA 99352
Attn: R. E. Rhoads

E. I. du Pont de Nemours & Co.
Savannah River Plant
Aiken, SC 29801
Attn: S. Petrie

Electric Power Research Institute
P. O. Box 10412
Palo Alto, CA 94304
Attn: T. Marston

Exxon Nuclear Co., Inc.
777 106th Avenue NE
Bellevue, WA 98004
Attn: J. H. Nordahl

General Atomic Company
P. O. Box 81608
San Diego, CA 92138
Attn: R. M. Burgoyne

General Electric Company
175 Curtner Avenue
San Jose, CA 95125
Attn: C. Davis

Hanford Engineering Development Lab
P. O. Box 1970
Richland, WA 99352
Attn: P. R. Clements

Lawrence Livermore Lab (1)
P. O. Box 808
Livermore, CA 94550
Attn: K. C. MacMillan, L333
R. T. Langland, MS, L-90

Los Alamos Scientific Lab (1)
P. O. Box 1663
Los Alamos, NM 87545
Attn: H. J. Fullbright III, M-1
T. Butler, WX-8
David Smith

Nuclear Assurance Corporation
24 Executive Park West
Atlanta, GA 30329
Attn: C. Johnson

Nuclear Packaging, Inc.
1733 South Fawcett
Tacoma, WA 98402
Attn: J. D. Simchuk

Oak Ridge National Laboratory
P. O. Box X
Oak Ridge, TN 37830
Attn: L. B. Shappert

J. L. Ridihalgh, President
Ridihalgh & Associates
2122 Iuka Avenue
Columbus, OH 43201

Rockwell International (1)
Atomics International Division
Rocky Flats Plant
P. O. Box 464
Golden, CO 80401
Attn: R. W. Skulski
W. D. Stump
B. Cross

DISTRIBUTION (Cont'd.)
Unlimited Release

Southwest Research Institute
P. O. Drawer 28510
San Antonio, TX 78228
Attn: R. A. Baker

Stearns-Roger, Inc.
P. O. Box 5888
Denver, CO 80217
Attn: W. H. Brinkman

Transnuclear, Inc.
One North Broadway
White Plains, NY 10601
Attn: L. Macklin

U. S. Department of Energy (6)
Advanced Nuclear System
MS B107
Washington, DC 20545
Attn: F. Dieringer
S. Meyers
M. J. Lawrence
R. B. Chitwood
R. Staker
F. Falci

U. S. Department of Energy (3)
Albuquerque Operations Office
P. O. Box 5400
Albuquerque, Nm 87185
Attn: E. C. Hardin, Jr.
K. Carlson
R. Y. Lowrey

U. S. Department of Energy
FFTF Project Office
P. O. Box 550
Richland, WA 99352
Attn: John Patterson

U. S. Department of Energy
Office of Military
Division of Research, Development
and Testing
Washington, DC 20545
Attn: Paul O. Mathews, Research Branch

Applied Technical Services (5)
Marietta, GA
Attn: H. S. Pearson

U. S. Department of Transportation
MTB (DMT-221)
400 7th St. SW
Washington, DC 20590
Attn: R. Rawl

U. S. Nuclear Regulatory Commission (3)
Washington, DC 20555
Attn: W. R. Lahs
C. E. MacDonald
D. R. Hopkins

Westinghouse Electric Corp.
P. O. Box W
Oak Ridge, TN 37830
Attn: J. E. Rutenber

Westinghouse Hanford Co. (1)
HEDL, P. O. Box 1970, 300 Area
MS-WA 132
Richland, WA 93352
Attn: T. E. Michaels
E. A. Proudfoot
J. C. Spanner

1725 J. A. Anderson
4400 A. W. Snyder
Attn: D. J. McCloskey, 4410
J. V. Walker, 4420
J. A. Reuscher, 4450
4440 G. R. Otey
Attn: M. Berman, 4441
D. A. Dahlgren, 4443
4442 W. A. VonReisemann
Attn: L. J. Klamerus, 4442
4500 E. H. Beckner
Attn: W. D. Weart, 4510
R. W. Lynch, 4530
M. L. Kramm, 4540
4550 R. M. Jefferson
Attn: R. E. Luna, 4551

DISTRIBUTION (Cont'd.)
Unlimited Release

4552 R. E. Pope (26)
4552 G. C. Allen
4552 T. A. Duffey
4552 R. G. Eakes
4552 G. H. Lamoreaux
4552 J. M. Ortman
4552 L. E. Romesberg
4552 S. H. Sutherland
4552 A. A. Trujillo
4552 M. G. Vigil
4552 H. R. Yoshimura
5000 J. K. Galt
Attn: F. L. Vook, 5100
O. E. Jones, 5500
D. B. Shuster, 5600
5520 T. B. Lane
Attn: L. W. Davison, 5521
T. G. Priddy, 5522
R. C. Reuter, 5523
R. T. Othmer, 5524
5522 R. A. May
5800 R. S. Claassen
Attn: R. E. Whan, 5820
N. J. Magnani, 5840
5810 R. G. Kepler
Attn: L. A. Harrah, 5811
J. G. Curro, 5813
5830 M. J. Davis
Attn: R. W. Rohde, 5832
J. L. Jellison, 5833
D. M. Mattox, 5834
J. L. Ledman, 5836
5835 C. H. Karnes
5835 R. E. Blose
5835 H. J. Rack (4)
3141 T. L. Werner, (5)
3151 W. L. Garner (3)
For: DOE/TIC
(Unlimited Release)
4550 TTC Master File
4551 TTC Library (7)
8266 E. A. Aas
DOE/TIC (25)
(R. P. Campbell, 3154-3)

Org.	Bldg.	Name	Rec'd by*	Org.	Bldg.	Name	Rec'd by*

*Recipient must initial on classified documents.